Inst. Phys. Conf. Ser. No 114: Section 1
Paper presented at RIS 90, Varese, Italy, 16–21 September 1990

Detection of very rare isotopes by collinear resonance ionization of accelerated atoms

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ABSTRACT: Results are presented of the work on the development of a method for detecting ultrarare radioactive isotopes, based on the collinear isotope-selective laser ionization of atoms in a fast beam. The rare isotope ^{3}He was detected at a selectivity of 10^9, and the isotope-selective ionization of krypton atoms was realized. A scheme is suggested for detecting the rare radioactive isotopes ^{85}Kr and ^{81}Kr.

1. INTRODUCTION

One of the problems in laser spectroscopy is to develop a detector for very rare radioactive isotopes contained in samples together with the abundant stable isotopes of the same elements (Letokhov 1981, 1987; Hurst 1988). The solution of this problem will help to solve many a problem in nuclear physics, geochemistry, environment protection, detecting past events, and so on.

The paper describes a fairly universal method of detecting rare isotopes, based on the collinear isotope-selective laser ionization of fast isotopic atoms in a beam. The idea of the method (Kudryavtsev, Letokhov 1982) is as follows. The atoms of all isotopes in the substance of interest are ionized and accelerated to an energy of eU_A in an ion source and then turned to neutral in a charge-exchange cell. The ions that failed to turn to neutral atoms are extracted from the beam by means of a filtering capacitor. The atoms of the isotope to be detected are then raised to a Rydberg state in an isotope-selective fashion in a field-free region. The excited atoms are ionized in the electric field of an ionizer, and the ions thus produced are deflected onto a detector. Since in the field-free region all the isotopic atoms of the same energy move with different velocities, there occurs an additional isotope shift in collinear excitation. The magnitude of this shift is given by

$$\Delta\nu^{is} = \pm(\nu_0/c)(2eU_A)^{1/2}[1/(M_1)^{1/2} - 1/(M_2)^{1/2}],$$

where M_1 and M_2 are the masses of two isotopes, ν_0 is the atomic transition frequency, and c is the velocity of light. The plus and minus signs correspond to the laser light directions with and against the atomic beam, respectively. The solid lines in Fig. 1 show the additional isotope shift (for $\Delta M = 1$) as a function of the isotope mass for the energies eU_A equal to 10 and 100 keV. The circles indicate the natural isotope shifts for resonance transitions of various elements. It can be seen that at an

energy of a mere 10 keV the additional isotope shift for atoms in the middle of the periodic table is 100 times their natural shift. It is also essential that an artificial isotope shift exists in any transition of any element. Another important feature of the method is that the Doppler absorption linewidth in the collinear excitation of a fast atomic beam is reduced by a factor of $2(eU_A/kT)^{1/2}$ and can be commensurable with the radiative linewidth (Kaufman, 1976). This allows one to make a narrow-band laser radiation interact with all the atoms of the isotope being detected in the fast beam and achieve their highly selective excitation. The method makes it possible to effect a stepwise excitation to Rydberg states of high-ionization-potential atoms by means of the existing dye lasers, provided the atoms possess high-lying metastable states to be populated in the course of charge exchange. It is also necessary that the ionization potential of the atoms contained in the charge-exchange-cell be close to the electron binding energy in the metastable states.

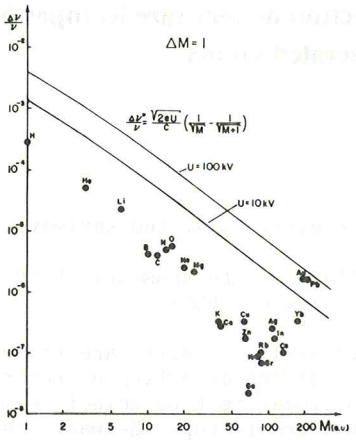

Fig. 1. Relative isotope shift of resonance lines of various elements (circles) and kinematic isotope shift of accelerated atoms 10 and 100 keV in energy as a function of atomic mass (solid curves) for $\Delta M = 1$

2. DETECTION OF THE ISOTOPE ^{3}He

The concentration of the rare isotope ^{3}He relative to that of the abundant isotope ^{4}He in various objects varies over a wide range. To illustrate, the relative concentration of ^{3}He in the atmosphere is equal to 1.4×10^{-6} and remains the same throughout the globe; it is 10^{-7}-10^{-8} in the commercial-grade helium gas obtained from natural gas deposits and comes to 10^{-9}-10^{-10} in some minerals.

The experiments on detecting the rare isotope ^{3}He (Kudryavtsev, Petrunin 1988) have shown that the capabilities of the technique are limited by the background noise signal due to collisions between the accelerated atoms of the abundant isotope ^{4}He and residual gas molecules. At a residual gas pressure in the vacuum system of 10^{-7} mm Hg, the level of the background noise signal corresponded to a ^{3}He concentration of 10^{-6}.

In the present work, to combat the background noise, use was made of an additional time-of-flight separation of the accelerated isotopic atoms in a pulsed beam. This made it possible to suppress greatly the noise signal and detect reliably the rare isotope ^{3}He in samples with its relative concentration as low as 10^{-9}.

The experimental setup is shown schematically in Fig. 2*a*. A beam of helium ions accelerated to an energy of eU_A = 3.9 keV is produced by a hot-cathode ion source and then focused with a single electrostatic lens and deflected by means of a deflector d_1 to free it from the neutral component. The deflected beam is then made to pass through a plane

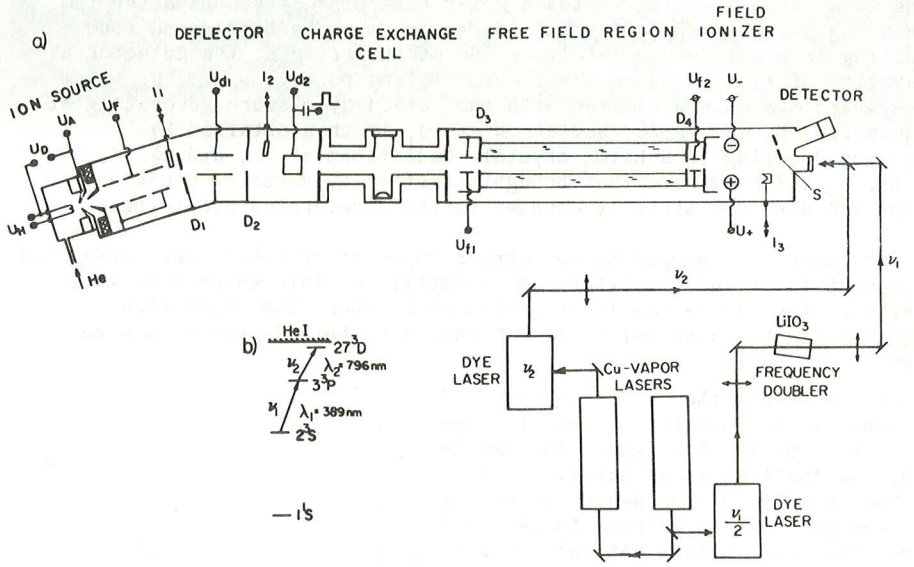

Fig. 2. (*a*) Schematic diagram of the experimental setup and (*b*) energy level diagram of the helium atom

capacitor d_2 used as an ion beam modulator. By applying a voltage of U_{d2} = 50 V across the capacitor d_2, the ions can be completely deflected from the axis of the setup. Modulation is effected by means of short square pulses returning the ion beam to the axis. The time of flight from the modulator to the detector is 4.5 μs for ^{3}He and 5.2 μs for ^{4}He. With the modulating pulse duration τ_p being as short as 0.5 μs, the isotopes are separated completely.

Downstream of the modulator, the ion beam enters a charge exchange cell containing potassium vapor at a temperature of 160°C, in which around 40% of the ions exchange their charge to become neutral atoms, some 3/4 of the atoms being produced in the 2^3S triplet state. The ions that failed to exchange their charge and the atoms produced in highly excited states are extracted from the beam by the electric field (9 kV/cm in strength) of a filtering capacitor f_1. Neutral atoms enter a 1 m long field-free region where they undergo two-step laser excitation from the 2^3S metastable state state to the 27^3D Rydberg state via the 3^3P intermediate level. Excitation is effected when a group of ^{3}He atoms (at τ_p = 0.5. μs, the group is 25 cm long) is at the exit from the field- free region (diaphragm D_4 2 mm in diameter). The diagram of the energy levels of He used in excitation is presented in Fig. 2*b*.

The Rydberg atoms entering the electric field of the two cylinders ⊕ and ⊖ (usually U_{f2} = 0) are ionized and deflected by the same field onto an adjustable slit S with a secondary electron multiplier behind. Such ionization of the Rydberg atoms by the transverse electric field features dispersion with respect to n, i.e., the signals from different n^3D states are observed at different potentials U_+ and U_- of the cylinders.

The $^3He^+$ ions are detected during a time interval of τ_{det} = 0.6 μs by means of a gated counting system a delay time of τ_d = 4.5 μs after the modulating pulse is applied to the capacitor d_2. The background ions resulting from collisions involving ^4He atoms arrive at the detector a delay time of t_d = 5.2 μs after the modulating pulse.
The Rydberg states are excited with two pulsed dye lasers generating at frequencies ν_2 and $\nu_1/2$. Radiation at ν_1 is then obtained by frequency doubling in a $LiIO_3$ crystal. Radiations at ν_1 and ν_2 (λ_1 = 389 nm, λ_2 = 796 nm) are then brought together by means of a dielectric mirror and directed strictly counter to the accelerated atomic beam.

The dye lasers are pumped by two copper-vapor laser tubes, one being used as an oscillator and the other, as an amplifier. This ensures reliable locking of the dye lasers in step with each other. The repetition frequency of the laser pulses is 8.6 kHz, and their duration is some 18 ns.

Figure 3 presents the counting rate of ^3He ions as a function of the isotope concentration in the gas. As can be seen, the function is a straight line, and the results of our measuremenets of ^3He isotopic concentrations in air and commercial-grade helium gas are in full agreement with the literature data. The counting rate of photoions of a rare isotope at a concentration of C is $N = I\tau_p f \eta C$, where I is the continuous atomic beam current, τ_p is the beam current pulse duration, f is the laser pulse repetition frequency, and η is the efficiency of exciting the atoms and detecting the photoions produced. For f = 8.6 kHz, $I = 2.3\times10^{11}$ s^{-1}, τ = 0.5 μs, $C = 10^{-6}$, and η = 0.01, the ^3He photoion counting rate is N = 10 counts per second. The background noise signal corresponds to a ^3He concentration of 10^{-9} at a vacuum of 10^{-7} Torr in the system. The efficiency of detecting the rare isotope atoms, determined as the ratio between the counted number N of atoms to the total number of atoms in the beam, equal to $C\times I$, amounted to 4.3×10^{-5}. The detection efficiency can be improved tenfold, to amount to 4×10^{-4}, by enhancing the laser pulse energy. The background noise signal in that case will correspond to a ^3He concentration of 10^{-10}.

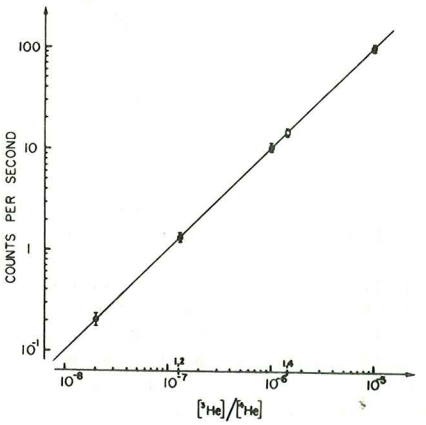

Fig. 3. Photoion signal from ^3He as a function of ^3He concentration. The square indicates helium gas extracted from the atmosphere

3. ISOTOPE-SELECTIVE IONIZATION OF KRYPTON ATOMS

The detection of the rare radioactive krypton isotopes ^{81}Kr ($T_{1/2} = 2.1\times10^5$ years, $C = 5\times10^{-13}$) and ^{85}Kr ($T_{1/2}$ = 10.7 years, $C = 5\times10^{-11}$) is of interest in environment protection, as well as in the study of hydrological processes occurring on a time scale of hundreds of thousands of years.

At present, there exists a method for detecting rare krypton isotopes, based on a combination of a repeated filtration of the rare isotope of interest by means of a mass filter with subsequent laser resonance photoionization detection of Kr atoms (Thonnard *et al.* 1987).

Our experiment on the isotope-selective ionization of krypton atoms was conducted on the setup described above. Metastable krypton atoms in the $1S_5$ state were prepared by quasiresonant charge exchange between krypton ions and potassium atoms (Fig. 4). The first-step wide-band dye laser radiation of $\Delta\nu_1 = 1$ cm^{-1} propagating counter to the atomic beam raised all the krypton isotopes from the $1S_5$ to the $2P_6$ level, $\lambda_1 = 7602$ Å. The laser was pumped with the yellow line of a copper vapor laser. The second-step radiation ν_2 was the green line of the copper laser, $\lambda_2 = 5106$ Å. This line resonated with the krypton atoms on the $2P_6$–$24d[3/2]_2$ transition, the atoms being 2.7 keV in energy and the light propagating in the same direction as the atomic beam. To reduce the bandwidth $\Delta\nu_2$ to some 0.03 cm^{-1}, use was made of two Fabry-Perot etalons 10 and 30 mm in free spectral range. To tune to resonance with different isotopes in the second-step transition, the atomic beam was retarded by applying a decelerating voltage across the charge-exchange cell, the ionizer voltage being corrected in step. The experimental ionization spectrum of krypton isotopes is shown in Fig. 5*a*. A 1-GHz change in frequency corresponds to a 31-eV change in the atomic energy. Figure 5*b* shows the theoretical spectrum of all even Kr isotopes and the odd isotope ^{85}Kr, along with their relative concentrations. The width of the spectrum obtained is determined by the laser linewidth $\Delta\nu_2$ and amounts to 850 MHz.

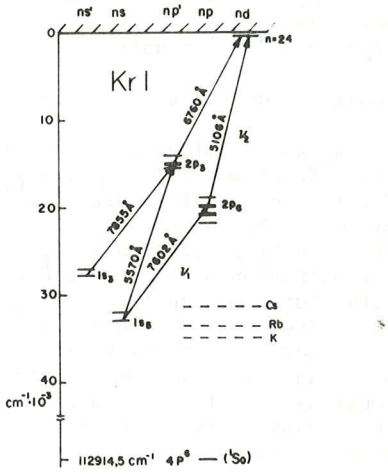

Fig. 4. Energy level diagram of the Kr atom

As in the case of the isotope He, rare krypton isotopes can be detected in conjunction with a preliminary mass separation, so that the collisional background noise can be reduced. To implement the method, use should be made of a pulsed narrow-band radiation in both excitation steps, which can be obtained by amplifying the radiation of a single-mode CW dye laser in a pulsed amplifier pumped by a copper vapor laser. Let us estimate the selectivity and sensitivity of the method using the $1S_5 \dashrightarrow 2P_3 \dashrightarrow 24d$ transitions whose hyperfine splitting constant for the ^{85}Kr isotope are known. The theoretical absorption spectrum for the first-step transition $1S_5 \dashrightarrow 2P_3$, $\lambda_1 = 5570$ Å is shown in Fig. 6. The natural isotope shift in this transition is 50 MHz. When the atoms are accelerated to 10 keV, the isotope shift increases to 1.7 GHz. Because of the hyperfine level splitting of ^{85}Kr ($I = 9/2$), its strongest line, $F = 13/2 \dashrightarrow F = 11/2$ (extreme right) is 1.9 GHz distant from the nearest ^{86}Kr line. The laser excitation selectivity in this step is $S_1 = 2.4\times10^5$ (at $\Gamma = 3$ MHz). The selectivity S_2 in the second step will be of the same order of magnitude as S_1, and the selectivity of the two-step excitation process, $S = S_1 \times S_2$, will be no less than 10^{10}. Should the preliminary mass separation ensure an enrichment of $S_M = 10^5$, the total selectivity will be high

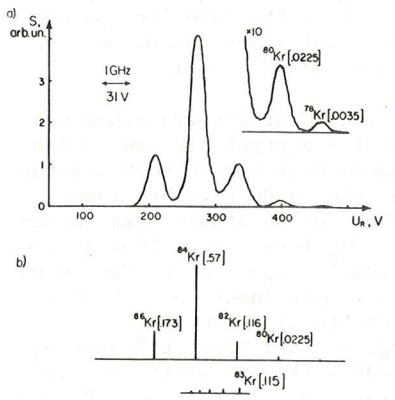

Fig. 5. (*a*) Photoion signal as a function of the decelerating voltage; (*b*) theoretical absorption spectrum of Kr isotopes for the $2P_6 \dashrightarrow 24d[3/2]$ transition

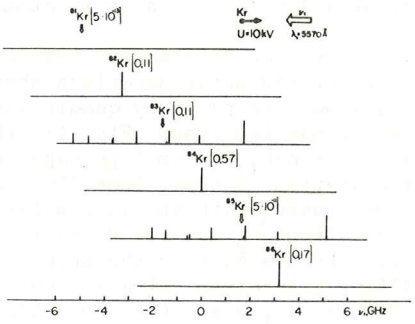

Fig. 6. Theoretical absorption spectrum of 10-keV isotopic Kr atoms for the $1S_5 \dashrightarrow 2P_3$ transition

enough to detect both ^{85}Kr and ^{81}Kr.

Let us now consider the sensitivity of the method. The counting rate for the ^{85}Kr atoms is $N = \eta_{c\text{-}e} \times \eta_{ex} \times \eta_{det} \times I_{Kr} \times \eta_{1S_5} \times C \times \tau_{det} \times f$, where $\eta_{c\text{-}e}$ = 0.5 is the charge-exchange efficiency, $\eta_{1S_5} = 5/12$ the proportion of atoms in the $1S_5$ state, $\eta_{ex} = 0.16$ the excitation efficiency, $\eta_{det} = 0.8$ the detection efficiency, I_{Kr} the total krypton ion current, C the concentration of the isotope being detected, $\tau_{det} = L/v = 7$ μs is the detection time equal to the time it takes for the atoms to traverse the field-free region, and $f = 10^4$ Hz is the laser pulse repetition frequency. The counting rate for ^{85}Kr at a concentration of 5×10^{-11} is 0.1 counts/s at an ion current of $I_{Kr} = 1.1\times10^{12}$ ions/s (180 nA). The counting rate for the background ions produced by collision is $N_{bg} = 10^{-3}$ counts/s at a vacuum of 10^{-9} Torr. Thus, the signal/noise ratio will be equal to 10^2.

The experimental results on the detection of ^{3}He and the estimates presented point to the possibility of using the collinear laser photoionoization method in conjunction with preliminary mass selection for detecting other isotopes (^{90}Sr, ^{3}H) of interest to environmental sciences.

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